The budget and partitioning of reactive nitrogen species in the Arctic stratosphere

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Volume mixing ratio profiles of numerous gases including NO, NO2, HNO3, HNO4, ClNO3, N2O5 and N2O were measured remotely from 8 to 38 km by the JPL MkIV FTIR solar absorption spectrometer during balloon flights from Fairbanks, Alaska (64.8N, 147.6W) on May 8 and July 8, 1997. The observed ratio of NOx (=NO+NO2) to NOy (total reactive nitrogen) is 10 to 30% greater than calculated by a steady state model using standard photochemistry and constrained by MkIV measurements of long lived precursors (e.g., H2O, CH4, CO and N2O) and SAGE II aerosol surface area. The persistence of this discrepancy to 38 km altitude suggests that processes involving aerosols, such as the reduction of HNO3 on the surface of soot particles, cannot be the sole explanation. Calculations using a 35% decrease for the rate of NO2+OH->HNO3 (the dominant sink of NOx in the Arctic summer stratosphere) compare well with MkIV observations of NOx/NOy and the individual NOy species at all altitudes. The good agreement between theory and observation of N2O5 suggests that the heterogeneous hydrolysis of N2O5, a minor sink of NOx during arctic summer, is handled correctly in the model. The MkIV NOy vs N2O relation agrees reasonably well with relations determined using the AER 2D model. In addition, comparison of the MkIV NOy vs N2O relation measured outside the vortex to ER-2 observations obtained inside the polar vortex on April 26, 1997 provides a context for inferring the degree of denitrification, which may have been as high as 5 ppbv during the 1996-97 Arctic winter.